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(54) **Silicone rubber composition for application as electrical insulation**

(57) A silicone rubber composition is disclosed for use as electrical insulation that cures into a highly water-resistant silicone rubber that has excellent electrical properties, and more particularly has excellent high-voltage electrical insulation properties. This silicone rubber composition comprises

- (A) polyorganosiloxane,
- (B) aluminum hydroxide powder surface treated

with an organosilane or organosilazane,

and

(C) curing agent.

Also, our composition is enhanced afforded by the optional addition of 1 to 200 weight parts of (D) a silica filler to the composition formed by combination of components (A) to (C).

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Description

This invention is a silicone rubber composition for application as electrical insulation (hereinafter EI silicone rubber compositions). This invention is particularly suited for application as electrical insulation because after its cure it exhibits an excellent water resistance and also excellent electrical properties after cure. Surprisingly, the invention has good volume resistivity, dielectric constant, dissipation factor, tracking resistance, arc resistance, and erosion resistance.

Compositions made by blending aluminum hydroxide powder into silicone rubber compositions are already known. It is also known that silicone rubbers with cure of these silicone rubber compositions [JP-B 5-12805 (1993) and JP-A 7-57574 (1995)]. However, these silicone rubber compositions contain large amounts of aluminum hydroxide powder, which is strongly water absorptive, and results in poor water resistance.

These compositions will also absorb moisture with the passage of time, leading to a decline in their electrical insulating properties. Hence, they are not fully acceptable as an EI silicone rubber composition for high-voltage use.

The present invention was achieved as a result of extensive research by the present inventors directed to solving the problems described above. In specific terms, the object of this invention is to provide a silicone rubber composition that cures into a highly water-resistant silicone rubber that has excellent electrical properties.

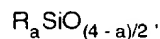
A silicone rubber composition is provided herein for application as electrical insulation that cures into a highly water-resistant silicone rubber that has excellent electrical properties. Particularly, it has excellent high-voltage electrical insulation properties. Our silicone rubber composition comprises

- (A) polyorganosiloxane,
- (B) aluminum hydroxide powder surface treated with an organosilane or organosilazane,
- and
- (C) curing agent.

Also, our composition is enhanced by the optional addition of 1 to 200 weight parts of (D), a silica filler, to the composition formed by combination of components (A) to (C).

The present invention is an EI silicone rubber composition comprising

- (A) 100 weight parts of polyorganosiloxane comprising at least 2 silicon-bonded alkenyl groups in each molecule and having the average formula



where R is selected from substituted or unsubstituted monovalent hydrocarbon groups and a has a value from 1.95 to 2.05,

(B) 1 to 300 weight parts of aluminum hydroxide powder surface-treated with an organosilane or organosilazane, and

(C) curing agent in a quantity sufficient to cure the composition.

The present EI silicone rubber composition can further comprise (D) 1 to 200 weight parts of silica filler.

The polyorganosiloxane (A) is described by the average formula $R_a SiO_{(4-a)/2}$. R in this formula is selected from substituted or unsubstituted monovalent hydrocarbon groups; and is exemplified by alkyl groups such as methyl, ethyl, and propyl; alkenyl groups such as vinyl, allyl, butenyl, and hexenyl; aryl groups such as phenyl; and 3,3,3-trifluoropropyl, 2-phenylethyl, and 2-cyanoethyl. Methyl preferably makes up at least 50 mole% of the R groups. The subscript a has a value from 1.95 to 2.05. Moreover, component (A) contains at least 2 silicon-bonded alkenyl groups in each molecule. The alkenyl can be bonded in terminal or non-terminal positions, and at both positions. The molecular structure of component (A) is straight chain or partially branched straight chain.

The viscosity of component (A) is not critical, but a viscosity at 25°C of from 100 to 20,000,000 mPa.s is preferred. Component (A) may be a homopolymer or copolymer or a mixture of such polymers. The units constituting component (A) are exemplified by the dimethylsiloxy, vinylmethylsiloxy, methylphenylsiloxy, and (3,3,3-trifluoropropyl)methylsiloxy units. The endblocking groups for component (A) are exemplified by trimethylsiloxy, silanol, vinyltrimethylsiloxy, and vinylmethylhydroxysiloxy. The instant polyorganosiloxane is exemplified by vinyltrimethylsiloxy-endblocked dimethylsiloxy-vinylmethylsiloxy copolymers, vinyltrimethylsiloxy-endblocked polydimethylsiloxy, silanol-endblocked dimethylsiloxy-vinylmethylsiloxy copolymers, vinylmethylhydroxysiloxy-endblocked dimethylsiloxy-vinylmethylsiloxy copolymers, and vinyltrimethylsiloxy-endblocked dimethylsiloxy-methylphenylsiloxy-vinylmethylsiloxy copolymers.

The surface-treated aluminum hydroxide powder (B) of the present composition has its surface treated with an

organosilane or organosilazane. It is the essential component for providing our silicone rubber composition with water resistance and electrical properties. The aluminum hydroxide powder component (B) has a particle size preferably from 0.1 to 100 μm , and more preferably from 0.1 to 50 μm . The organosilane which is used as a treating agent for component (B) is selected from methyltrimethoxysilane, methyltriethoxysilane, phenyltrimethoxysilane, ethyltrimethoxysilane, n-propyltrimethoxysilane, vinyltrimethoxysilane, allyltrimethoxysilane, butenyltrimethoxysilane, hexenyltrimethoxysilane, γ -methacryloxypropyltrimethoxysilane, dimethyldimethoxysilane, dimethyldiethoxysilane, diphenyldimethoxysilane, tetraethoxysilane, and the partial hydrolyzates of the preceding. The organosilazane which can be used as a treating agent for component (B) is exemplified by hexamethyldisilazane or divinyltetramethyldisilazane.

Component (B) is prepared by treating the surface of the aluminum hydroxide powder with the organosilane or organosilazane by adding the powder and treating agent organosilane or organosilazane to a mixer, and then blending. The treatment temperature during this procedure is preferably from 50 to 200°C, and more preferably from 80 to 180°C. This preparation can also be carried out during formation of the silicone rubber base compound by first adding the powder and treating agent, to component (A), and by then mixing while heating. The ratio of the organosilane or organosilazane to the aluminum hydroxide powder during preparation is preferably from 0.1 to 30 weight%.

Component (B) is added to the present composition at from 1 to 300 weight parts and preferably at from 30 to 200 weight parts, in each case per 100 weight parts of component (A). The addition of less than 30 weight parts component (B) leads to inadequate results in terms of electrical properties, such as tracking resistance and arc resistance. The addition of more than 200 weight parts component (B) causes a substantial embrittlement of the cured product and prevents good rubber properties.

The curing agent (C) used in the present composition cures the composition. Organoperoxides are a typical example of this component and include benzoyl peroxide, tert-butyl perbenzoate, 2,4-dichlorobenzoyl peroxide, dicumyl peroxide, and 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane.

Another example of curing agent (C) is the combination of a hydrosilylation reaction catalyst and polyorganohydrogensiloxane comprising at least 3 silicon-bonded hydrogen atoms in each molecule. The polyorganohydrogensiloxane is a crosslinker component. In this embodiment, our claimed composition undergoes crosslinking and curing as a result of the addition reaction of the silicon-bonded hydrogen in the polyorganohydrogensiloxane of component (C) to the silicon-bonded alkenyl in component (A) in the presence of the hydrosilylation reaction catalyst. The polyorganohydrogensiloxane must contain at least 3 silicon-bonded hydrogen atoms in each molecule. The non-hydrogen silicon-bonded organic groups in the polyorganohydrogensiloxane are exemplified by alkyl groups such as methyl, ethyl, and propyl; aryl groups such as phenyl or tolyl; and substituted alkyl groups such as 3,3,3-trifluoropropyl or 3-chloropropyl.

This polyorganohydrogensiloxane can have a straight-chain, branch-containing straight-chain, cyclic, or network molecular structure. While its viscosity is not critical, its viscosity at 25°C is preferably from 3 to 10,000 mPa.s. This component is added in a quantity that provides a value from 0.5 : 1 to 20 : 1, and preferably from 1 : 1 to 3 : 1 for the ratio of the number of moles of silicon-bonded hydrogen to the silicon-bonded alkenyl in the composition. The present composition will not undergo an acceptable cure when this molar ratio has a value below 0.5. A value in excess of 20 can cause the production of gaseous hydrogen and undesirable foaming.

The hydrosilylation reaction catalyst used herein as the second embodiment of curing agent (C) is a cure-accelerating catalyst. Preferred among such catalysts are platinum metal, platinum compounds, and compositions whose principal component is a platinum compound, in each case the particular species manifests a catalytic activity that accelerates the hydrosilylation reaction. The platinum catalysts are exemplified by chloroplatinic acid, alcohol-modified chloroplatinic acid, platinum/diketone complexes, platinum/olefin complexes, chloroplatinic acid/alkenylsiloxane complexes, and these species supported on a carrier such as alumina or carbon black. This component is added in a catalytic quantity, generally at from 1 to 150 weight parts, and preferably at from 5 to 100 weight parts, in each case as platinum metal for each 1,000,000 weight parts of component (A).

The silica filler (D), an optional component herein, is a reinforcing filler. This component is added when higher levels of mechanical strength are required of our silicone rubber composition. The silica (D) is exemplified by reinforcing silicas such as dry-process silica, for example, fumed silica; wet-process silicas, for example, precipitated silica; and by semi-reinforcing silica such as quartz or diatomaceous earth. Preferred as this component are those silica fillers having a particle sizes of $\leq 50 \mu\text{m}$ and a specific surface area of $\geq 50 \text{ m}^2/\text{g}$. Even more preferred are silicas whose surfaces have been hydrophobicized by treatment with an organosilicon compound such as an organosilane, organosilazane, or organosiloxane oligomer.

High mechanical strength of the cured composition is not achieved when too little of component (D) is added, and a loss of fluidity occurs when too much is added. As a result, component (D) is added at from 1 to 200 weight parts, and more preferably at from 1 to 60 weight parts, in each case per 100 weight parts of component (A). When component (D) is added, the total addition of components (B) and (D) is preferably from 30 to 300 weight parts per 100 weight parts component (A).

Thus the present composition comprises the components (A), (B), and (C), or the components (A), (B), (C), and

(D). Insofar as the object of our invention is not impaired, the claimed composition may also contain those additives heretofore known for use with silicone rubber compositions. For example, additives may be added including non-reinforcing fillers, pigments, heat stabilizers, flame retardants, internal release agents, and surface-treatment agents. The non-reinforcing fillers are exemplified by diatomaceous earth, quartz, mica, aluminum oxide, and titanium oxide. The pigments are exemplified by carbon black and iron oxide red; while the heat stabilizers are exemplified by rare earth oxides, rare earth hydroxides, cerium silanates, and the cerium salts of fatty acids.

The present composition are simply prepared by mixing the above-described components (A), (B), and (C), or the components (A), (B), (C), and (D) to homogeneity. These components are mixed by using those conventional means heretofore known in the art. For example, kneader mixers, twin-screw continuous compounding extruders, and two-roll mills are all useful herein. When an organoperoxide is used as the curing agent (first embodiment), the present composition is cured by heating to a temperature at or above the decomposition temperature of the organoperoxide, for example, 130 to 200°C. When the combination of a hydrosilylation reaction catalyst and polyorganohydrogensiloxane is used (second embodiment), the composition is conveniently cured by heating to 70 to 200°C. The method for molding our present silicone rubber composition is selected according to the particular molding method, such as compression molding or extrusion molding.

The present composition cures into highly water-resistant products that evidence low water absorption and excellent electrical properties. These products also have good high-voltage electrical insulating properties. Thus, the claimed silicone rubber composition is well suited for example, for the preparation of silicone rubber electrical insulation for outdoor use.

The invention is explained below through working examples, in which "parts" means "weight parts" and the values reported for viscosity were measured at 25°C. The electrical properties were measured in the examples using the following methods.

Measurement of the electrical properties of the silicone rubber composition was performed on samples press-vulcanized for 10 minutes at 170°C to mold a 2 mm-thick silicone rubber sheet. The volume resistivity, dielectric constant, and dissipation factor were measured by the methods provided in JIS C 2123, "Test Methods for Silicone Rubber Compounds". The volume resistivity was measured using a high resistance meter (High Resistance Meter 4339A from the Hewlett-Packard TM Company) and the dielectric constant and dissipation factor were measured using an automatic instrument for loss measurement in dielectrics (TR-1100 Model Automatic Loss Measurement Instrument for Dielectrics from Andoh Denki Kabushiki Kaisha, 110 Hz).

The anti-tracking test was run in accordance with the method of IEC Publication 587 using a model HAT-520 from Hitachi Kasei Kogyo Kabushiki Kaisha. The test voltage was 3.5 kV. The evaluation A referenced in the tables refers to the time required until the current flowing in a high-voltage circuit passing through the test specimen exceeded 60 mA. The evaluation B referenced in the tables refers to the time for tracking to reach a mark placed on the surface of the test specimen at a position 25 mm from the lower electrode.

Measurement of the water resistance of the silicone rubber composition was made on samples press-vulcanized for 10 minutes at 170°C to mold a 2 mm-thick silicone rubber sheet. This sheet was dipped in deionized water at room temperature for 100 hours, and then withdrawn; and the electrical properties were measured on the resulting sample. The % water absorption was also measured from the weight change in the sheet as determined from its weight pre- and post-immersion.

Example 1. A silicone rubber base compound was prepared by combining the following and mixing to homogeneity in a kneader mixer: 100 parts of vinyl dimethylsiloxyl-terminated dimethylsiloxane-vinyl methylsiloxane copolymer gum, containing 99.87 mole% of dimethylsiloxyl units and 0.13 mole% of vinyl methylsiloxyl units and 160 parts of aluminum hydroxide powder whose surface had been treated with vinyltrimethoxysilane (Higilite TM H42STV from Showa Denko Kabushiki Kaisha, average particle size = 1 µm). 0.8 part of a 50 weight% silicone oil paste masterbatch of 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane (curing agent) was mixed to homogeneity on a two-roll mill into 100 parts of the above silicone rubber base to give a curable EI silicone rubber composition. This silicone rubber composition was press-vulcanized for 10 minutes at 170°C to mold a 2 mm-thick silicone rubber sheet, on which the electrical properties and water resistance were measured. These measurement results are reported in Table 1.

Example 2. A silicone rubber base compound was prepared by combining the following and mixing to homogeneity in a kneader mixer: 100 parts of vinyl dimethylsiloxyl-terminated dimethylsiloxane-vinyl methylsiloxane copolymer gum, containing 99.87 mole% of dimethylsiloxyl units and 0.13 mole% of vinyl methylsiloxyl units; and 160 parts of aluminum hydroxide powder whose surface had been treated with γ-methacryloxypropyltrimethoxysilane (Higilite TM H320ST, average particle size = 8 µm). 0.8 part of a 50 weight% silicone oil paste masterbatch of 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane (curing agent) was mixed to homogeneity on a two-roll mill into 100 parts of the above silicone rubber base also to give a curable EI silicone rubber composition. This silicone rubber composition was press-vulcanized for 10 minutes at 170°C to mold a 2 mm-thick silicone rubber sheet, on which the electrical properties and water resistance were measured. These results are also reported in Table 1.

The values of Table 1 for the electrical properties and water resistance of the silicone rubber composition prepared

above were measured as in Example 1.

Comparative Example 1. A silicone rubber composition was prepared as in Example 1, but in this case using an untreated aluminum hydroxide powder (Higilite TM H42M, average particle size = 1 μm) instead of the Higilite H42STV of Example 1.

The properties of this silicone rubber composition were measured as in Example 1, and the results are given in Table 1.

Example 3. The following were mixed to homogeneity in a kneader mixer into 100 parts of vinyltrimethylsiloxy-endblocked dimethylsiloxane-vinylmethylsiloxane copolymer gum, containing 99.87 mole% dimethylsiloxy units and 0.13 mole% vinylmethylsiloxy units: 30 parts, surface-treated fumed silica with a specific surface area of 120 m^2/g (Aerosil R972 from Nippon Aerosil TM Kabushiki Kaisha) and 3 parts of silanol-endblocked polydimethylsiloxane with a viscosity of 30 mPa.s. This was followed by the addition of 160 parts of aluminum hydroxide powder (surface pre-treated with methyltrimethoxysilane) with mixing at room temperature. The addition to 100 parts of this base of curing agent as described in Example 1 afforded a curable silicone rubber composition. The methyltrimethoxysilane-treated aluminum hydroxide powder used in this case had been prepared by adding 1 part of methyltrimethoxysilane to 100 parts of an untreated aluminum hydroxide powder (Higilite TM H42M) and mixing in a mixer for 3 hours at 120°C.

The electrical properties and water resistance of this silicone rubber composition were then measured as in Example 1, and the results are given in Table 2.

Comparative Example 2. The following were mixed to homogeneity in a kneader mixer into 100 parts of vinyltrimethylsiloxy-endblocked dimethylsiloxane-vinylmethylsiloxane copolymer gum, containing 99.87 of mole% of dimethylsiloxy units and 0.13 mole% of vinylmethylsiloxy units: 30 weight parts fumed silica with a specific surface area of 200 m^2/g and 12 parts of silanol-endblocked polydimethylsiloxane with a viscosity of 30 mPa.s as surface-treatment agent. Heating at 175°C for 30 minutes then gave a silicone rubber base. 160 parts of untreated aluminum hydroxide powder (Higilite TM H42M, average particle size = 1 μm) were then mixed to homogeneity at room temperature into this silicone rubber base. The addition to 100 parts of this silicone rubber base of curing agent, as described in Example 1, then afforded a curable EI silicone rubber composition. The electrical properties and water resistance of this silicone rubber composition were measured as in Example 1, and the results are enumerated in Table 2.

Example 4. The following were mixed to homogeneity into 100 parts of vinyltrimethylsiloxy-endblocked polydimethylsiloxane with a viscosity of 10,000 mPa.s (silicon-bonded vinyl content = 0.14 weight%): 30 parts of fumed silica with a specific surface area of 200 m^2/g , 150 parts of untreated aluminum hydroxide powder (Higilite TM H42M) and, as surface-treatment agent, 10 parts of hexamethyldisilazane and 3 parts of water. This was followed by mixing for an additional 2 hours at 170°C in a vacuum to give a silicone rubber base compound. A curable EI silicone rubber composition was then prepared by mixing this base compound to homogeneity with 1.4 parts of trimethylsiloxy-endblocked dimethylsiloxane-methylhydrogensiloxane copolymer with a silicon-bonded hydrogen content of 0.7 weight% and, as hydrosilylation reaction catalyst, sufficient 3 weight% isopropanolic chloroplatinic acid solution to provide 10 ppm of platinum metal. The electrical properties and water resistance of the cured EI silicone rubber composition were measured as in Example 1, and these results are shown in Table 3.

Table 1

	Example 1	Example 2	Comparative Example 1
initial			
volume resistivity ($\Omega\text{-cm}$)	1.2×10^{14}	1.5×10^{14}	1.9×10^{14}
dielectric constant	4.4	4.0	4.0
dissipation factor	1.4×10^{-2}	9.0×10^{-3}	1.2×10^{-2}
tracking resistance			
evaluation A (min)	≥ 360	≥ 360	≥ 360
evaluation B (min)	≥ 360	≥ 360	≥ 360
after dipping in water for 100 hours			
volume resistivity ($\Omega\text{-cm}$)	1.1×10^{14}	9.5×10^{13}	2.4×10^{10}
dielectric constant	4.8	4.5	7.8
dissipation factor	2.2×10^{-2}	1.5×10^{-2}	2.5×10^{-1}
water absorption (%)	0.2	0.25	1.9

Table 2

	Example 3	Comparative Example 2
initial		
volume resistivity (Ω -cm)	1.9×10^{14}	2.0×10^{14}
dielectric constant	4.4	4.2
dissipation factor	1.1×10^{-2}	2.0×10^{-2}
tracking resistance		
evaluation A (min)	≥ 360	≥ 360
evaluation B (min)	≥ 360	≥ 360
after dipping in water for 100 hours		
volume resistivity (Ω -cm)	7.0×10^{13}	2.1×10^{10}
dielectric constant	4.7	7.4
dissipation factor	3.3×10^{-2}	2.08×10^{-1}
water absorption (%)	0.28	1.5

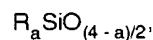
Table 3

	Example 4
initial	
volume resistivity (Ω -cm)	7.5×10^{14}
dielectric constant	3.5
dissipation factor	9.3×10^{-3}
tracking resistance	
evaluation A (min)	≥ 360
evaluation B (min)	≥ 360
after dipping in water for 100 hours	
volume resistivity (Ω -cm)	2.2×10^{14}
dielectric constant	3.8
dissipation factor	1.2×10^{-2}
water absorption (%)	0.15

Claims

1. A silicone rubber composition comprising

(A) 100 weight parts of polyorganosiloxane comprising at least 2 silicon-bonded alkenyl groups in each molecule and having the average formula



where R is selected from substituted or unsubstituted monovalent hydrocarbon groups and a has a value of from 1.95 to 2.05,

(B) 1 to 300 weight parts of aluminum hydroxide powder surface-treated with an organosilane or organosilazane,

and

(C) curing agent in a quantity sufficient to cure the composition.

2. A composition according to Claim 1, where the polyorganosiloxane has a viscosity at 25°C of from 100 to 20,000,000 mPa.s.

3. A composition according to Claim 2, where the polyorganosiloxane is selected from vinyldimethylsiloxy-endblocked polydimethylsiloxanes or vinyldimethylsiloxy-endblocked dimethylsiloxane-vinylmethylsiloxane copolymer.
4. A composition according to Claim 1, where the surface-treated aluminum hydroxide powder has a particle size of from 0.1 to 50 μm .
5. A composition according to Claim 1, where the surface treated aluminum hydroxide powder is surface treated with an organosilane selected from vinyltrimethoxysilane or methyltrimethoxysilane.
6. A composition according to Claim 1, where the surface treated aluminum hydroxide powder is surface treated with an organosilazane selected from hexamethyldisilazane or divinyltetramethyldisilazane.
7. A composition according to Claims 5 or 6, where the aluminum hydroxide powder is surface treated with from 0.1 to 30 weight percent of the organosilane or organosilazane.
8. A composition according to Claim 1, where the curing agent is an organoperoxide.
9. A composition according to Claim 1, where the curing agent is a combination of a hydrosilylation reaction catalyst and a polyorganohydrogensiloxane comprising at least 3 silicon-bonded hydrogen atoms in each molecule.
10. A composition according to Claim 1 further comprising (D) 1 to 200 weight parts of a silica filler.
11. A composition according to Claim 10 where the silica is a fumed silica having a specific surface area greater than or equal to 50 m^2/g .
12. A composition according to Claim 11, where the surface treated silica filler is provided by treatment with vinyltrimethoxysilane or methyltrimethoxysilane.



European Patent
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EUROPEAN SEARCH REPORT

Application Number
EP 97 30 3556

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
E	EP 0 787 772 A (DOW CORNING TORAY SILICONE CO. INC.) 6 August 1997 * claims 1-7 *	1,3-5,7,8	C08K9/06 C08L83/07
A	EP 0 645 779 A (NGK INSULATORS LTD ;DOW CORNING TORAY SILICONE (JP)) 29 March 1995 * claims 1,2 *	1	
D	& JP 07 057 574 A (TORAY DOW CORNING SILICONE CO LTD) 3 March 1995		
A	US 4 822 830 A (ADKINS JAMES R) 18 April 1989 * claims 1-16 *	1	
A	PATENT ABSTRACTS OF JAPAN vol. 017, no. 236 (C-1057), 13 May 1993 & JP 04 366170 A (SHIN ETSU CHEM CO LTD), 18 December 1992, * abstract *	1	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C08K C08L
The present search report has been drawn up for all claims			
Place of search MUNICH		Date of completion of the search 12 August 1997	Examiner Hoffmann, K
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